SIZEABLE DYNAMIC PROTON POLARIZATIONS*

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By extending previous work[1,2] to higher fields and frequencies, we have produced nuclear polarizations as large as 51 % for the protons in the waters of hydration in single crystals of lanthanum magnesium nitrate, $La_2Mg_3(NO_3)_{12}\cdot 24\ H_2O$, in which 1 % of La^{3+} has been replaced by paramagnetic Nd^{3+}. The method[3] is reviewed in Fig. 1, which shows the energy levels of a Nd^{3+} ion of effective spin $S=\frac{1}{2}$ and a proton of spin $I=\frac{1}{2}$ in a magnetic field H. One observes the usual electron spin resonance transitions a at microwave frequency ν_e, as well as nuclear resonance transitions b at $\nu_n \sim 10^{-3}\ \nu_e$. Because of small admixtures of states by the electron-nuclear dipolar coupling, it is also possible to induce "forbidden" transitions c and d by the applied microwave field H_1. If, e.g., c is strongly induced, the relative populations N of states $(-\frac{1}{2},-\frac{1}{2})$ and $(\frac{1}{2},\frac{1}{2})$ become equal to unity, while the electron spin-lattice relaxation maintains the populations of state $(\frac{1}{2},-\frac{1}{2})$ at $\exp(-h\nu_e/kT)$ and $(-\frac{1}{2},\frac{1}{2})$ at $\exp(+h\nu_e/kT)$; T is the crystal temperature, maintained by a bath of liquid helium. The nuclear polarization, defined by

$$p \equiv [N(M_I=\tfrac{1}{2}) - N(M_I=-\tfrac{1}{2})]/[N(M_I=\tfrac{1}{2}) + N(M_I=-\tfrac{1}{2})],$$

thus becomes ideally $p_i = \tanh(h\nu_e/2kT) \approx h\nu_e/2kT$.

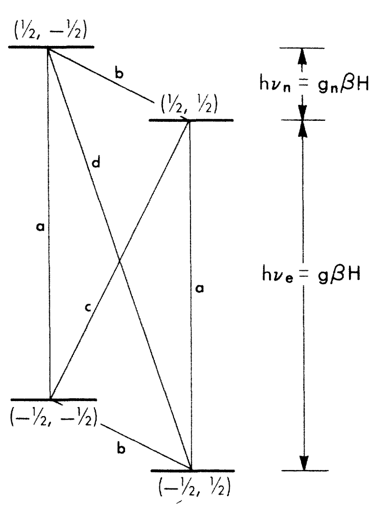

FIG. 1. Energy levels of an electron spin and a nuclear spin in weak dipolar coupling and in a magnetic field H. The states are labelled by the high-field quantum numbers (M_S, M_I).

This dynamic value is larger by ν_e/ν_n than the static value $p_0 = h\nu_n/2kT$. Similarly, saturation of d gives an enhanced polarization, but of opposite sign. Saturation of a reduces p to zero.

In practice the actual polarization enhancement E is found to be less than the ideal value ν_e/ν_n, because transitions a, c, and d are not completely resolved, and also because nuclear spin-lattice relaxation due to extraneous electron spins may not be negligible. To obtain large absolute values of the dynamic polarization $p=p_0E$, we have tried to do the following: (1) Make p_0 as large as possible by operating in a high H field. (2) Maximize E by making $H\nu_n/\nu_e$ large compared to the electron resonance linewidth, ΔH, so that a, c, and d are resolved; this also requires high fields. (3) Carefully eliminate paramagnetic impurities in the crystal other than the desired Nd^{3+} ions.

Lanthanum magnesium nitrate crystals are fairly suitable for dynamic polarization of protons since they are highly hydrogenous, and furthermore all rare-earth sites are magnetically equivalent. We grow them from aqueous solution in a desiccator at 0°C, using 99.997 % purity La. To avoid hfs lines, Nd enriched to 98.5 % even isotopes is used. The g factor of Nd^{3+} is anisotropic ($g_\perp = 2.70$, $g_\parallel = 0.36$), and this allows a variation of the operating field H (for a fixed frequency ν_e), through variation of the angle θ between H and the crystal z axis. The linewidth ΔH is also anisotropic[2]: At $\nu_e = 35$ kMc/sec, $\theta = 90°$, $H = 9.3$ kOe, and $\Delta H \approx 4.5$ Oe; at $\theta = 30°$, $H = 18$ kOe, and $\Delta H \approx 9$ Oe. The resolution of a, c, and d is unfortunately not very dependent on θ. At high fields Nd^{3+} has a considerably narrower linewidth than Ce^{3+}, used earlier at lower fields.[1,3]

Our dynamic polarization apparatus for use at $H \sim 20$ kOe, $\nu_e \sim 50$ kMc/sec, and $T \sim 1.5$°K is shown schematically in Fig. 2. The sample crystal, a flat hexagonal plate ~1 cm in diameter and ~2 mm thick, is placed in a nuclear resonance coil contained in a cylindrical microwave cavity with a volume of 3 cm^3. The coil is connected to a simple Q-meter type[4] of magnetic-resonance detector, which has the virtue of linearity. Since the crystal is larger than the wavelength of the microwaves, we use a high-mode tunable cavity,

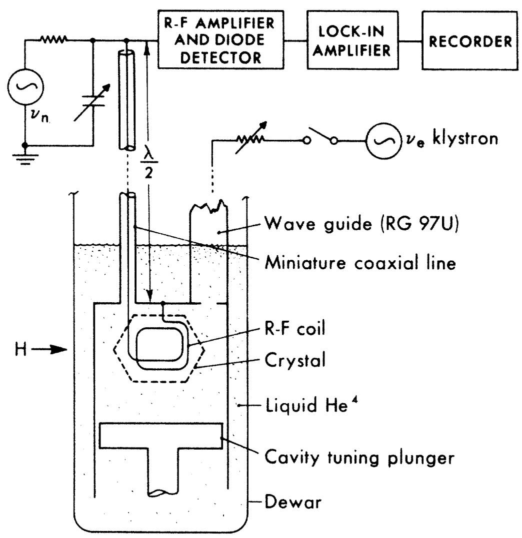

FIG. 2. Schematic diagram of proton-polarization apparatus.

which may be swept through several different resonant modes during the time the forbidden transitions are being induced; this assures that the microwave H_1 field will, on the average, irradiate the entire crystal uniformly. The cavity is coupled via a thin-wall waveguide, attenuator, and microwave switch to a klystron of fixed frequency ν_e.

Experiments are performed as follows: At thermal equilibrium at the bath temperature T, a nuclear resonance signal at ν_n is recorded. Then the microwaves are switched on and the H field set to a value to induce transition c (or d). After a steady state is reached (~5 minutes), the microwaves are switched off and the nuclear resonance is immediately recorded and found to be enhanced by the factor E. This enhanced proton polarization $p = Eh\nu_n/2kT$ is then observed to decay approximately exponentially to the thermal equilibrium value in a characteristic time T_{1n}. Some results are given in Table I, along with calculated values of the theoretical ideal polarization p_i. The largest polarization obtained, $p = 51\,\%$, is significantly greater than values previously reported, and indicates that even larger polarizations will be obtained at still higher fields and frequencies. The microwave power required to saturate a forbidden transition is ~20 mW for a crystal volume ~0.1 cm^3. One could probably produce a 50 % proton polarization in crystal volumes of several cm^3, which may be useful as polarized targets for high-energy nuclear scattering experiments.

A polarization of 51 % in a field $H_i = 20.1$ kOe corresponds to a proton spin temperature $T_i \approx 0.004$°K, so that a final spin temperature $T_f = H_f T_i / H_i$ in the microdegree range may be reached upon demagnetization to a low field H_f in times short compared to the proton spin-lattice relaxation times T_{1n}. We have demagnetized to a final proton spin temperature $T_f \approx 90$ microdegrees at $H_f \approx 500$ Oe, where we find $T_{1n} \approx 125$ sec. We observe that $T_{1n} \propto H^2$ in this region, so that lower temperatures could be reached and maintained only for increasingly shorter times.

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[1]O. S. Leifson and C. D. Jeffries, Phys. Rev. 122, 1781 (1961).

[2]P. L. Scott, thesis, University of California, Berkeley, California, 1961 (unpublished).

[3]The polarization of nuclei by inducing "forbidden" transitions which simultaneously flip nuclear and electron spins is a modification of the Overhauser effect and has been used to orient nuclei in hfs contact interaction with electron spins [C. D. Jeffries, Phys. Rev. 106, 164 (1957)]. For the case of weak dipolar coupling, a prototype experiment was done first for nuclear-nuclear systems [A. Abragam and W. G. Proctor, Compt. rend. 246, 2253 (1958); N. Bloembergen and P. P. Sorokin, Phys. Rev. 110, 865 (1958)], and then extended to electron-nuclear systems [E. Erb, J. L. Motchane, and J. Uebersfeld, Compt. rend. 246, 2121,

Table I. Results of dynamic proton-polarization experiments on crystals of $La_2Mg_3(NO_3)_{12}\cdot 24\ H_2O$ containing 1 % Nd enriched to 98.5 % even isotopes.

ν_n (Mc/sec)	ν_e (kMc/sec)	H (kOe)	T (°K)	E meas.	p meas.	p_i calc.	T_{1n} (min)
75	35	17.7	2.07	300	26 %	39 %	14
86.4	50	20.3	2.1	400	39 %	51 %	12
85.4	50	20.1	1.35	340	51 %	71 %	20

3050 (1958)]. Proton-polarization experiments in cerium magnesium nitrate have been reported by M. Abraham, M. A. H. McCausland, and F. N. H. Robinson, Phys. Rev. Letters 2, 449 (1959); and by M. Borghini and A. Abragam, Suppl. Helv. Phys. Acta, VI, 143 (1961).

[4]J. Hatton and B. V. Rollin, Proc. Roy. Soc. (London) A199, 222 (1949).

NEW METHOD FOR PRODUCING AND ANALYZING LINEARLY POLARIZED GAMMA-RAY BEAMS

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In this note we propose a new method for the production and the analysis of a polarized beam of high-energy gamma rays. The method is based on the interference effects which are observable in high-energy electron pair production on crystals. As a consequence of these the absorption rate (inverse of the mean free path) of very high-energy photons in crystal matter depends on their linear polarization. This suggests the possibility of using a thick crystal for the polarization and analysis of high-energy gamma rays. The polarization is effected by preferential absorption of the unwanted polarization component, and the analysis by transmission measurements, as in the case of a polaroid filter for visible light. Extensive theoretical work on the interference effects of high-energy electrodynamic processes in crystals has been done by Überall,[1] and refined experiments have given results which are in excellent agreement with the theory.[2]

Another method for the production of linearly polarized gamma rays by means of interference effects in a crystal is based on bremsstrahlung.[3] In this case the theory is also in excellent agreement with the experimental results obtained with electrons of ~1 GeV.[4]

As a polarimeter the device we propose is perhaps unique in the high-energy region, where the application of other methods based on the angular distribution in pair production and elastic photoproduction of π^0 on nuclei of zero spin[5] requires very difficult experiments. A unique characteristic of the device is that, since it is both a polarizer and an analyzer, one can build two or more of them and cross-calibrate them with each other. The polarizing power of the device can therefore be directly measured.

The absorption of high-energy photons is mainly due to electron pair production, a process which is already known to give interference effects.[1,2] Let us consider the case of a cubic crystal, where the momentum $\vec{k}$ of the incoming gamma rays is in the (001) plane and makes a small angle α with the (110) axis. The (001) plane is then a symmetry plane. We find that the total cross section for pair production depends in this situation on the linear polarization of the gamma rays.

Let us denote by $\Sigma^{\parallel}$ and $\Sigma^{\perp}$ the total cross sections per unit volume of the crystal for gamma rays which are linearly polarized in the (001) plane and orthogonal to it. The two polarization components will be absorbed with different mean free paths; i.e., after having penetrated a thickness x of the crystal the intensities of the two components will be reduced according to[6]

$$I^{\parallel}(x) = I^{\parallel}(0)\exp[-\Sigma^{\parallel}x],$$
$$I^{\perp}(x) = I^{\perp}(0)\exp[-\Sigma^{\perp}x]. \quad (1)$$

If the beam was originally unpolarized $[I^{\parallel}(0) = I^{\perp}(0)]$, we now have a polarization:

$$P(x) = [I^{\parallel}(x) - I^{\perp}(x)]/[I^{\parallel}(x) + I^{\perp}(x)]$$
$$= \tanh[\tfrac{1}{2}x(\Sigma^{\perp} - \Sigma^{\parallel})]. \quad (2)$$

From Eq. (2) one can see that this method could, in principle, produce any degree of polarization, with an appropriate choice of the thickness x. This is achieved with a loss of the original intensity which can be expressed in terms of the polarization $P(x)$ and of a param-